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(54) Title: SELF-FOAMING MICROEMULSION CLEANING COMPOSITIONS

(57) Abstract

The invention is a cleaning composition in the form of a microemulsion comprising a self-foaming agent in an effective amount to form a foam after the composition is dispensed on a surface, at least one cosurfactant, at least one surfactant, and the balance water, wherein the composition is a stable, self-foaming microemulsion having a sub-atmospheric vapor pressure at 22 °C and capable of being dispensed as a liquid from a non-pressurized package.

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SELF-FOAMING MICROEMULSION CLEANING COMPOSITIONS

The present invention relates to household cleaning products which foam after dispensing.

5 Household cleaning products have been provided to consumers by manufacturers in a variety of forms including especially solids and liquids which are commonly diluted in use and rinsed away, and sprays which are commonly applied directly to the surface or article to be
10 cleaned and subsequently rinsed or wiped away. Sprays are particularly popular as a product for cleaning hard surfaces such as spray bathroom cleaners used for showers enclosures, tubs, tile, sinks and the like; all purpose spray cleaners used to clean appliances, counters, kitchen
15 sinks, and the like; glass cleaners used for windows, mirrors and the like, and mold and mildew and disinfectant cleaners. Sprays are also popular as a product for pretreating fabrics prior to laundering and as rug and carpet cleaners. Sprays have the advantage that they
20 permit the broad application of a relatively small amount of a liquid cleaning composition to a surface or article to be cleaned. The application of a spray product is easily directed toward an article or surface, however it is often difficult to distinguish areas to which the spray
25 has been applied from areas which have not yet been sprayed.

Ordinarily, household cleaning products foam during the process of cleaning and in general consumers desire a product which foams well, provided that the foam
30 is easily removed or dissipated from the cleaned article or surface. Thus, liquid cleaning products such as dish washing liquids and liquid laundry detergents are optimized to exhibit good foaming. These products are diluted in a bath before use and are used in situations

where the foam is easily rinsed away from the cleaned article.

Foaming is also desired in the case of liquid or spray products used to clean hard surfaces. With spray bathroom cleaners there is usually ample opportunity for rinsing. However, there is often less opportunity for effective rinsing of all purpose spray cleaners and glass cleaners. Spray products which exhibit good foaming as they are used tend to leave residual foam when wiped from the cleaned article. As a result, spray products used to clean hard surfaces, especially all purpose spray cleaners and glass cleaners, are usually low foaming compositions which tend to leave less residual foam when wiped clean.

Manufacturers of household cleaning products have resorted to a number of means to improve their foaming characteristics. Aerosol containers have found particular application as a means of dispensing a product as a foam. However, aerosol products have suffered from negative public perception due to their perceived adverse environmental effects. Further, aerosol foams provide a product which is dispensed in the form of a foam rather than a liquid product which foams as it cleans.

Post-foaming compositions are known in personal care products particularly as post foaming shave gels. These post foaming compositions have not found application in household cleaning products for a number of practical reasons. In general, these compositions, typified by post-foaming shave gels, must be dispensed from expensive compartmented aerosol packages such as the package shown in Figure 4 of US Patent No. 3,541,581 and the piston package described in US Patent No. 4,913,323. This is due both to the super atmospheric vapor pressure of the compositions which necessitates pressurized packaging and to the thick, non-fluid consistency of the gel which

cannot be adequately exhausted from a non compartmented pressurized package.

Self-leveling, self-foaming liquid compositions which incorporate a dual purpose pressure agent used to
5 both dispense the liquid from a non-compartmented barrier package and to foam the liquid have been described in the art. Such compositions nevertheless exhibit super-atmospheric vapor pressure and must thus be contained in and dispensed from a pressurized package of a barrier
10 material.

Other cleansing compositions having self-foaming properties contain as self-foaming agents organic liquids whose vapor pressure is sub-atmospheric. However, such compositions exhibit viscosities which make them
15 unsuitable for dispensing from a non-aerosol spray device. Further, such compositions employ high levels of surfactants which are difficult to rinse from a cleaned surface or article and unnecessarily add to product cost.

Other cleansing compositions contain sub-atmospheric organic liquid compounds having self-foaming
20 properties and relatively high surfactant levels. These compositions use water soluble organic gums as viscosity modifiers to give the shampoo a viscosity in the range of 1000 to 20,000 centistokes at 5°C or the compositions are
25 in the form of gels. In general, liquids with such high viscosities are unsuitable for dispensing as a non-aerosol spray since the high viscosity prevents the liquids from being satisfactorily atomized. Additionally, cleaning compositions containing high levels of surfactants can be
30 difficult to rinse from the cleaned surface and add to the cost of such products.

Another reference teaches the use of a water-insoluble volatile organic liquid, pentane, in an "aqueous saponaceous composition" having a sub-atmospheric vapor

pressure to produce compositions which foam spontaneously when the composition is spread out into a thin layer. The composition could be dispensed from an atomizer. However, the composition was not stable in that the pentane did not
5 remain in solution. The composition required agitation or shaking immediately prior to use in order to distribute the pentane throughout the composition. These compositions also required higher surfactant concentrations to obtain their foaming effects.

10 Cleaning product compositions which incorporate a perfume as the essential hydrocarbon in the form of a microemulsion have been described in the art. However, these compositions do not self-foam.

 Liquid crystal detergent compositions which
15 incorporate low vapor pressure hydrocarbon solvents for the cleaning of oily soils are described in the art. However, the hydrocarbon solvents required in these compositions are insufficiently volatile to produce a composition that is self-foaming. Other compositions
20 which lack a self-foaming agent exhibit no self-foaming property although the compositions may foam when agitated or when sprayed through a sprayer designed to impart foaming to a liquid.

 There is therefore a need for improved hard
25 surface cleaning products which foam well after dispensing but are easily rinsed or wiped from a cleaned surface or article and which do not tend to leave residual foam after wiping.

 In the application of cleaning products to an
30 article or surface to be cleaned it is desirable to apply a relatively small amount of product broadly so as to effectively cover the surface which is to be cleaned. To this end it is desirable that the area to which product has already been applied be visible. There is therefore a

need for hard surface cleaning products in a form which allows the user to apply a relatively small amount of product broadly and to easily see the area to which product has been applied.

5 It is also desirable that the cleaning product remain in the area to which it has been applied until rinsed, wiped or otherwise removed from the article or surface. In the case of horizontal surfaces liquid and spray products tend to remain where applied. However, in
10 the case of inclined and especially vertical surfaces liquids and sprays tend to run and drip.

To counteract this tendency, hard surface cleaning products in the form of thickened liquids have been developed and marketed. Such thickened products are
15 more effectively retained on an article or surface to be cleaned. However, thickened cleaning products usually require greater effort and care in rinsing because of their tendency to be more effectively retained on the article or surface. Further, such thickened liquids do
20 not lend themselves to application in the form of a spray since they are difficult to atomize and project sufficiently. There is therefore a need for liquid cleaning compositions which cling well to vertical surfaces when applied as a liquid or a spray and which
25 self-foam after application.

Summary of the Invention

In one aspect, this invention is a post-foaming composition comprising:

(a) water

30 (b) from about a 0.5 to about 10 weight percent of a water immiscible compound or mixture thereof, the water immiscible compound(s) having a vapor pressure which satisfies the relationship

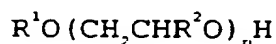
$$3 \text{ psi} < \sum_{i=1}^{i=y} X_i P_i < 14.7 \text{ psi.}$$

wherein y represents the number of water immiscible compounds in component (c), x_i represents the mole fraction of a water immiscible compound i in component (c), and P_i represents the vapor pressure of said water-immiscible compound i at 22° C;

(c) from about 0.2 to about 20 weight percent of a surfactant; and

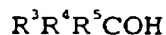
(d) from about 0.1 to about 20 weight percent of a cosurfactant selected from the group consisting of

1) an alkylene glycol ether of the formula



where R^1 is C_1 - C_6 alkyl, R^2 is H or methyl and n is a number from 1 to 3,

2) an alcohol of the formula



where R^3 , R^4 and R^5 are each either H or C_1 - C_6 alkyl provided that the total number of carbon atoms in R^3 , R^4 and R^5 is 3 to 7, and

3) mono- and di-carboxylic acids having an equivalent weight of up to 88;

the relative amounts of components (a)-(d) being selected such that the composition is in the form of a liquid oil-in-water microemulsion.

The composition of this invention may be dispensed as a non-foamed liquid spray. Some period of time after dispensing, which is usually in the order of

about 1 to 20 seconds, the sprayed composition foams. In this way, the surface onto which the composition is sprayed can be readily identified. Furthermore, the composition of this invention is storage stable for a
5 period of 30 days or more and can be packaged in non-pressurized containers. The composition also clings well to vertical surfaces and provides for excellent cleaning.

In a second aspect, this invention is a container having a non-pressurized reservoir and an
10 opening, said reservoir containing the post-foaming composition of the first aspect of the invention, said opening being fitted with a dispensing apparatus which is adapted to be actuated from a closed to an open position wherein when said dispensing apparatus is actuated to an
15 open position, it imparts shear to said post-foaming composition and dispenses it from the container in the form of a liquid, non-foamed spray.

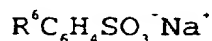
Detailed Description of the Invention

Water is an essential component of the
20 composition of this invention. Water is necessary to provide both the desired foaming and the lathering qualities and is used in quantities sufficient to obtain a liquid of desired viscosity. Water from any source, including for example, tap water, distilled water, and
25 deionized water is suitable for use in the present invention provided any impurities and additives in the water do not interfere with the functionality of the other components. The amount of water in the composition is ultimately determined by the amount of the other essential
30 and optional ingredients included in the composition. Preferably, deionized water is used. Preferably, from about 60 percent to about 98 percent by weight of the composition is water. More preferably, from about 80 percent to about 98% by weight of the total composition is
35 water.

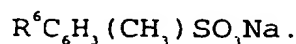
The composition also contains a surfactant, which performs several functions. It aids in stabilizing the microemulsion and helps to stabilize the foam which forms after the composition is dispensed. Surfactants
5 which are useful herein have a alkylaryl, alkyl or alkenyl hydrocarbon chain, of from about 10-32 carbon atoms, preferably an alkyl chain of about 10-30 carbon atoms.

In general, the efficiency of a surfactant in forming microemulsion compositions of the present
10 invention increases as the number of carbon atoms in the alkyl chain increases. Surprisingly, in compositions of the present invention it is normally preferred to employ a surfactant with an alkyl chain containing more than 12 carbon atoms. This contrasts with the normal practice of
15 formulating conventional foaming compositions such as shampoos where an increase in the alkyl chain length of the surfactant beyond 12 carbon atoms results in lower amounts of foaming.

Useful surfactants used in compositions of the
20 present invention include for example, anionic, cationic, nonionic, amphoteric, or zwitterionic surfactants or mixtures thereof. Mixtures of an anionic and a nonionic surfactant are preferred. Useful anionic surfactants include for example, metal salts of alkylbenzene
25 sulfonates and alkyltoluene sulfonates. Examples of useful anionic surfactants include for example, those having the formula

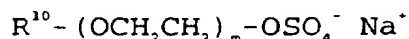


where R^6 is a linear or branched, inertly substituted, C_{12} -
30 C_{30} hydrocarbon, preferably a C_{12} - C_{30} alkyl and alkyltoluene sulfonates having the formula



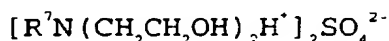
Inert substituent include hydroxyl groups, unsaturated sites, and the like which do not destroy the surface active character of the compound.

Preferred anionic surfactants include those in which the R⁶ group contains 20 to 24 carbon atoms. Other useful anionic surfactants include alkyl ethe sulfate surfactants of formula:



where R¹⁰ is an alkyl group of 10-20 carbon atoms and m is a number from 0-12.

Preferred cationic surfactants used in compositions of the present invention include those represented by the formula



where R⁷ is a C₁₀₋₂₄ inertly substituted, linear or branched hydrocarbyl, preferably C₁₆₋₁₈ alkyl.

It is understood that many other surfactants and combinations of surfactants which are commercially available and known in the art may be used in the compositions of the present invention without departing from the principles taught herein. A compilation of such commercially available surfactants is found in McCutcheon's Detergents and Emulsifiers which is incorporated herein by reference.

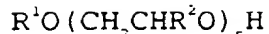
The surfactant must also be compatible with the other materials employed in the composition including the oxygen-containing cosurfactant, and any optional ingredients such as salts, hydrotropes, antimicrobial agents, acids, fragrance, colorant and the like. By "compatible", it is meant that the materials do not engage in an unwanted chemical reaction with each other which

adversely affects their ability to perform the desired function in the composition, and that the materials will not phase separate in the composition.

5 The amount of surfactant used is selected in conjunction with the amounts and types of the other ingredients so that a stable oil-in-water microemulsion is formed. The microemulsion is clear or slightly hazy. Generally, the surfactant is present in compositions of the present invention in an amount of from about 0.2% to
10 about 20% by weight of the composition. Preferably, the surfactant is present in compositions of the present invention in an amount of from about 0.5% to about 10% by weight. More preferably, the surfactant is present in the compositions in an amount of from about 0.5% to about 7%
15 by weight. The surfactant may be used in excess of that amount needed to form a stable microemulsion, but great excesses are not desired as that increases cost and viscosity.

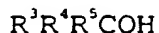
Another essential ingredient in compositions of
20 the present invention is a cosurfactant. The cosurfactant lowers the interfacial tension between the oil phase and the water phase and allows formation of microemulsion systems. The cosurfactant functions in combination with the surfactant to form a microemulsion incorporating the
25 desired amount and type of self-foaming agent. Compositions of the present invention may contain combinations of two or more cosurfactants. The cosurfactant may also function to aid in cleaning of resistant soils such as hard soap scum in the case of a
30 bathroom cleaner. It is also preferred to use a cosurfactant which has the lowest possible vapor pressure to minimize the content of volatile organic compounds in the composition as a whole.

Useful cosurfactants include alkylene glycol
35 ethers having the formula



where R^1 is C_1 - C_6 alkyl, R^2 is H or methyl and n is a number from 1 to about 3.

Other useful cosurfactants used in compositions
5 of the present invention either alone or in combination with the aforementioned cosurfactants include alcohols having the formula



where R^3 , R^4 and R^5 are each either H or C_1 - C_6 alkyl provided
10 that the total number of carbon atoms in R^3 , R^4 and R^5 is from 3 to about 9.

Other useful cosurfactants used in forming compositions of the present invention either alone or in combination with the aforementioned cosurfactants include
15 for example, mono- and di-carboxylic acids having an equivalent weight of up to about 88, such as acetic acid, adipic acid, and succinic acid.

More preferred cosurfactants include glycol ethers such as dipropyleneglycol n-butyl ether,
20 dipropylene glycol n-propyl ether, propyleneglycol n-butyl ether, propyleneglycol n-propyl ether, tripropyleneglycol methyl ether and mixtures thereof.

The amount of cosurfactant required in compositions of the present invention depends upon the
25 particular surfactant and water immiscible compound which are chosen for a particular composition. Generally, the cosurfactant is present in the composition in an amount of from about 0.1% to about 20%. Preferably, the cosurfactant is present in the composition an amount of
30 from about 0.5% to about 10%. More preferably, from about 1% to about 5% cosurfactant by weight is present in the compositions of the present invention.

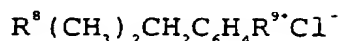
In addition to the aforementioned essential ingredients, the composition of the invention may contain other additives. Of particular importance are certain water soluble inorganic salts and hydrotropes. A water
5 soluble inorganic salt may be added to assist a particular combination of surfactant and cosurfactant in forming the microemulsion of the present invention. Salts may be added directly to the composition or formed in situ through the neutralization of an appropriate acidic
10 substance. Appropriate acidic substances include for example, citric acid, hydrochloric acid, and sulfuric acid. The acid may be neutralized with sodium hydroxide or another appropriate base. Preferred salts used in compositions of the present invention include for example,
15 sodium sulfate, sodium bisulfate, magnesium sulfate, sodium citrate, magnesium citrate, sodium carbonate, sodium chloride, and magnesium chloride. These salts, when used may constitute from about 0.1 to about 10% of the weight of the composition.

20 Hydrotropes are materials which are used in liquid detergent formulations to solubilize high concentrations of surfactant into an aqueous liquid composition and to suppress the formation of lyotropic liquid crystal phases in aqueous detergent compositions.
25 Since surfactants with alkyl chains containing higher numbers of carbon atoms are generally more susceptible to the formation of liquid crystal phases in aqueous solutions it may be useful to incorporate a hydrotrope into the compositions of the present invention to suppress
30 the formation of liquid crystals and promote the formation of a stable microemulsion. Generally, any compatible hydrotrope may be used in compositions of the present invention. Useful hydrotropes include for example, alkylaryl sulfonates with short alkyl chains such as
35 potassium toluene sulfonate, ammonium xylenesulfonate, sodium cumene sulfonate and medium chain alkane sulfonates

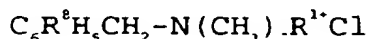
such as sodium primary octane sulfonate. Preferred hydrotropes have an aromatically bound strongly ionic group and no aliphatic hydrocarbon chain of greater than 8, preferably 4, most preferably 2 carbon atoms. A particularly preferred hydrotrope is sodium cumene sulfonate. When present, the hydrotrope advantageously constitutes about 0.1 to 5 weight % of the composition, on an active basis (i.e., excluding any water or solvent which is added with the hydrotrope).

Acids, other than those described above as cosurfactants, may be useful in compositions of the present invention. Acids are useful ingredients in many types of hard surface cleaning compositions including for example, bathroom and toilet bowl cleaners. Acids in the composition aid in the removal of hard water stains and soap scum. While no acid is required for the practice of the present invention, any convenient acid may optionally be used. Acids which are useful in compositions of the present invention include for example, organic acids such as citric acid and lactic acid, and inorganic acids such as sodium hydrogen sulfate and hydrochloric acid.

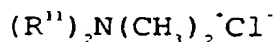
Antimicrobial agents may also be used in compositions of the present invention. Antimicrobial agents are commonly incorporated into hard surface cleaners to sanitize or disinfect surfaces to be cleaned. Generally, any compatible antimicrobial agent known to those skilled in the art may be used in compositions of the present invention. Useful antimicrobials include for example, quaternary ammonium compounds, including materials corresponding to the formulae



and



R_8 is a C_{10} - C_{18} carbon atom branched or linear hydrocarbon and R_9 is a lower alkyl, where R^9 is a C_{12} - C_{20} alkyl group, including, for example, a mixture of 60% C_{14} , 30% C_{16} , 5% C_{12} and 5% C_{18} or a mixture of R_1 is 68% C_{12} , 32% C_{14} and R^1 is hydrogen or a lower alkyl; and materials corresponding to the formula



where R^{11} is C_8 - C_{10} alkyl, and such phenolic antimicrobial agents as for example, ortho-phenylphenol and ortho-benzyl chlorophenol.

Compositions of the present invention may also contain other optional ingredients including perfumes and other water insoluble compounds not included among the aforementioned self foaming agents. When those optional ingredients are water immiscible these form part of the water immiscible compounds (b) and, together with any other water-immiscible compound, must satisfy the relationship set forth above.

Typical pH ranges for compositions of the present invention are from about pH 1 to about pH 13 depending on the intended cleaning application.

The compositions of the present invention are advantageously stable for a period of at least about 30 days at ambient temperatures, and preferably are stable for a like period over a temperature range of 0°C to 50°C.

The microemulsion of this invention can be prepared by combining the water and surfactant until the surfactant is dissolved. The cosurfactant is then combined with the surfactant mixture and mixed until visually homogeneous. The resulting solution is advantageously cooled to from about 0°C to about 10°C and the water immiscible compound(s) (which is preferably also

cooled to about 0°C to about 10°C) is added to the mixture and the composition is mixed. The composition is then transferred to an appropriate barrier container for storage.

5 Alternatively, all of the ingredients of a predetermined microemulsion composition, except the water immiscible compounds, may be combined in an intermediate as described above. An appropriate amount of this intermediate may be weighed into a container of barrier
10 material and an appropriate weight of cooled (0 to 10°C) water immiscible compound is added. The container is closed and the water immiscible compound(s) is incorporated into the composition with agitation.

 Compositions of the present invention may be
15 packaged in any convenient containers that provide a barrier to the self-foaming agent. Useful barrier materials include for example, polyethylene terephthalate (PET) and polyvinylchloride (PVC). The container preferably has a single non-pressurized reservoir in which
20 the product communicates with the head space. The reservoir has an opening to the outside which is fitted with a dispensing apparatus. The dispensing apparatus is advantageously made to be actuated between an open and closed position. Preferably, when the dispensing
25 apparatus is activated, it imparts a shear to the composition contained inside, so that the viscosity drops and it can be atomized and sprayed. Such dispensing apparatus are well known and include finger actuated atomizing spray valves and trigger sprayers. When in the
30 closed position, the dispensing means should provide a barrier to escape of the contents of the reservoir. The composition may be dispensed by any convenient means including for example, by pouring, by squeezing, or by spraying from a trigger sprayer. Preferably, the
35 composition is dispensed through a trigger sprayer, for

example, from a model T85NDB trigger sprayer available from Continental Sprayers.

5 The following examples are illustrative of the composition of the present invention and are not intended as limitations of this invention. Many apparent variations of which are possible without departing from the spirit and scope thereof. The stated amounts of the ingredients are parts by weight unless otherwise
10 indicated.

 The requisite amounts and types of the ingredients for the compositions of the following examples are predetermined by the process described hereinabove.

EXAMPLE 1

15 This example illustrates a microemulsion composition useful as a disinfectant cleaner.

 The surfactant is dissolved in water at room temperature and the ortho-phenylphenol is dissolved in the cosurfactant, again at room temperature. The two
20 solutions are blended together, mixed until homogeneous and then citric acid is added and dissolved. The mixture is then cooled to about 5°C. Isopentane is separately cooled to about 5°C and then is added to the mixture and mixed for about 2 minutes. The composition is transferred
25 to a PET bottle and sealed with a cap. The composition components are:

Component	Weight %
Sodium C _{11.5} alkylbenzene sulfonate ^a	8.33
Dipropyleneglycol-n-butyl ether ^b	4.17
Citric acid, anhydrous	2.00

Ortho-phenylphenol ^c	0.20
Isopentane (P at 22°C = 11.96 psi)	5.00
Water, deionized	to 100
$\Sigma_{i=1}^{i=n} X_i P_i$	11.96 psia

^a= Nacconol 90G, Stepan Company; ^b= Dowanol DPnB, The Dow Chemical Company; ^c= Dowicide 1, The Dow Chemical Company

EXAMPLE 2

5 This example illustrates a rapidly foaming microemulsion cleaning composition. The composition components are:

Component	Weight %
10 C ₂₀ -C ₂₄ alkylbenzene sulfonate, Na salt	0.75%
Dipropyleneglycol-n-butyl ether ^a	3.0
Propyleneglycol-n-butyl ether	0.98
Citric acid	2.0
15 Isopentane	3.5
Petroleum Distillates ^b	0.5
Sodium hydroxide	0.2
Water, deionized	to 100
$\Sigma_{i=1}^{i=n} X_i P_i$	11.03 psia

20 ^a= Dowanol DPnB, the Dow Chemical Company; ^b= NAPTHA SC 140, commercially available from Ashland Chemical. Its vapor pressure at 22°C is 0.019 psi.

25 The surfactant, cosurfactants and citric acid are mixed together at room temperature to form a solution and then cooled to about 5°C. The isopentane and the petroleum distillate are mixed together, cooled to about 5°C, added slowly to the surfactant solution and mixed

until clear. The composition is then stored in an appropriate barrier container.

EXAMPLE 3

Example 3 illustrates a rapidly foaming disinfectant cleaning composition. The composition components are:

Component	Weight %
10 Tridecylbenzene sulfonic acid ^a	5.20
Sodium cumene sulfonate (40% aq. sol.)	3.33
Dipropyleneglycol-n-propyl ether ^b	2.67
Citric acid, anhydrous	2.00
Sodium hydroxide	0.67
15 Ortho-phenylphenol ^c	0.20
Sodium sulfate	1.00
Isopentane	5.00
Water, deionized	to 100
20 $\sum_{i=1}^{i=y} X_i P_i$	11.96 psia
^a = Biosoft S130, Stepan Company; ^b = Dowanol, The Dow Chemical Company; ^c = Dowicide 1, The Dow Chemical Company	

The tridecylbenzene sulfonic acid is dissolved in water at room temperature. Sodium cumene sulfonate is added and mixed until dissolved. The ortho-phenylphenol is dissolved in the ether cosurfactant; the resulting solution is added to the surfactant solution and mixed. Sodium hydroxide is gradually added to obtain a pH of from between about 5 to about 9. Citric acid and sodium sulfate are added and the solution is mixed until they are dissolved. Ninety-five weight parts of this mixture are placed into a container. Isopentane is cooled to about 5°C

and then added to the mixture. The container is sealed and the mixture was blended to form a homogeneous microemulsion.

EXAMPLE 4

5 This example illustrates a slowly foaming disinfectant cleaning composition. The composition components are:

10	Component	Weight %
	Tridecylbenzene sulfonic acid ^a	5.20
	Sodium cumene sulfonate (40% aq. sol.)	3.33
	Dipropyleneglycol-n-butyl ether ^b	2.67
	Citric acid, anhydrous	2.00
15	Sodium hydroxide	0.67
	Ortho-phenylphenol ^c	0.20
	Sodium sulfate	2.00
	Isopentane (38.7 wt. %)/	5.00
	ISOPAR TM C ^d (61.3 wt. %)	
20	Water, deionized	to 100
	$\sum_{i=1}^{i=y} X_i P_i$	6.4 psia

^a= Biosoft S 130, Stepan Company; ^b= Dowanol DPnB, The Dow Chemical Company; ^c= Dowicide 1, The Dow Chemical Company; ^d= ISOPARTM C is a blend of isoparaffinic hydrocarbons primarily containing 2,2,4-trimethylpentane. It is commercially available from Exxon Chemicals. The vapor pressure of 2,2,4-trimethylpentane at 22°C is 0.8245 psia; this value is used in calculating $\sum_{i=1}^{i=y} X_i P_i$ in this example.

30 The isopentane and ISOPAR are blended and cooled to about 5°C. Otherwise, the composition is prepared as described in Example 3.

EXAMPLE 5

This example illustrates an acidic disinfectant cleaning composition. The composition components are:

5	Component	Weight %
	Sodium bisulfate ^a	2.00
	Di(hydroxyethyl)tallowamine ^b	6.00
	Cationic Antimicrobial ^c	3.00
10	Dipropyleneglycol-n-butyl ether ^d	3.00
	Citric acid, anhydrous	2.00
	Isopentane	5.00
	Water, deionized	to 100
15	$\sum_{i=1}^{15} X_i P_i$ psia	11.96

^a= from Jones Hamilton; ^b= Biosoft TA2, Stepan Chemical Company; ^c= BTC 2125M is a mixture of C₁₂₋₁₈ dimethylbenzyl ammonium chloride and C₁₂₋₁₈ dimethylethylbenzyl ammonium chloride commercially available from Stepan Chemical Company; ^d= Dowanol DPnB, The Dow Chemical Company.

The sodium bisulfate is dissolved in water, the dihydroxyethyl tallow amine is added and the solution is mixed until homogeneous. The antimicrobial, ether cosurfactant, and citric acid are added and mixed until dissolved. Ninety-five parts of this solution are transferred to a barrier container, cooled isopentane (about 5°C) is added, the container sealed, and the solution mixed until homogeneous.

EXAMPLE 6

This example illustrates a rapidly foaming cleaning composition. The composition components are:

	Component	Weight %
	Dodecylbenzene sulfonic acid ^a	3.24
	Dipropyleneglycol-n-butyl ether ^b	1.50
5	Magnesium oxide	0.20
	Citric acid, anhydrous	2.00
	Isopentane	5.00
	Water, deionized	to 100
10	$\sum_{i=1}^{12} X_i P_i$	11.96 psia
	a= Biosoft S100, Stepan Company; b= Dowanol DPnB, The Dow Chemical Company	

The surfactant, ether cosurfactant, magnesium
 15 oxide, and citric acid are sequentially dissolved in
 water. Isopentane is cooled to about 5°C, added to 95% by
 weight of the surfactant solution and mixed in a closed
 container until homogeneous.

EXAMPLE 7

20 This example illustrates a cleaning composition
 having a mixture of surfactants and a mixture of
 cosurfactants. The composition components are:

	Component	Weight %
25	Sodium C _{11.5} alkylbenzene sulfonate	1.00
	C ₂₀ -C ₂₄ alkyltoluenesulfonate, Na salt	0.75
	Dipropyleneglycol-n-propyl ether	2.00
	Propylene glycol-n-butyl ether	1.07
30	Citric acid	2.00
	NAPTHA SC-140	0.50
	Sodium hydroxide	0.66
	Isopentane	3.50
	Water, deionized	to 100
35	$\sum_{i=1}^{12} X_i P_i$	11.03 psia

The C₂₀₋₂₄ alkyltoluene sulfonate, sodium salt surfactant, propyleneglycol-n-butyl ether, and NAPTHA SC-140 are dissolved in 55% of the total amount of water. The sodium dodecylbenzene sulfonate, dipropyleneglycol-n-propyl ether, sodium hydroxide, and citric acid are added to the mixture, then mixed to form a solution and the remaining water is added. The solution is cooled to about 5°C and cooled isopentane (about 5°C) is added to the solution with mixing to form a microemulsion.

10 EXAMPLE 8

This example illustrates a cleaning composition that foams more slowly than the composition of Example 7. The composition components are:

15	Component	Weight %
	Sodium dodecylbenzene sulfonate	1.00
	C ₂₀₋₂₄ Alkyltoluenesulfonate, sodium salt	0.75
	Dipropyleneglycol-n-propyl ether	2.00
20	Propylene glycol-n-butyl ether	1.07
	Citric acid	2.00
	NAPTHA S-140	0.50
	Sodium hydroxide	0.86
	Isopentane (75 mole%)/ ISOPAR C (25 mole%)	3.50
25	Water, deionized	to 100
	$\sum_{i=1}^{15} X_i P_i$	9.18 psia

This composition is prepared as described in Example 7.

30 EXAMPLE 9

This example illustrates a microemulsion cleaning composition of the present invention having n-

pentane as the water immiscible compound. The composition components are:

5	Component	Weight %
	Sodium dodecylbenzene sulfonate	1.00
	C ₂₀₋₂₄ Alkyltoluenesulfonate, Na salt	0.75
	Dipropyleneglycol-n-propyl ether	2.00
	Propylene glycol-n-butyl ether	1.07
10	Citric acid	2.00
	NAPTHA S-140	0.50
	Sodium hydroxide	0.60
	n-Pentane	3.50
	Water, deionized	to 100
15	$\sum_{i=1}^{i=N} X_i P_i$	8.16 psia

This composition was prepared as described in Example 7.

20 EXAMPLE 10

This example illustrates an acidic disinfectant cleaning composition. The composition components are:

25	Component	Weight %
	Dodecyl/tridecylbenzene sulfonic acid	5.20
	Sodium cumene sulfonate (40% aq. sol.)	5.66
	Dipropyleneglycol-n-butyl ether	2.67
	Citric acid, anhydrous	2.00
30	Sodium hydroxide	0.67
	Ortho-phenylphenol	0.20
	Sodium sulfate	1.00
	Isopentane (65.45 weight %)	
	/ISOPAR C (34.55 weight %)	5.00
35	Water, deionized	to 100

$$\sum_{i=1}^{i=9} X_i P_i$$

9.17 psia

The surfactant and sodium cumene sulfonate are dissolved sequentially in the water. Ortho-phenylphenol is dissolved into the ether cosurfactant and the cosurfactant is added to the surfactant solution with continued agitation. Sodium hydroxide is slowly added to the mixture to obtain a pH of from about 5 to about 9. Citric acid and sodium sulfate are added and the solution is mixed at room temperature until homogeneous. The isopentane/ISOPAR mixture is cooled to about 5°C and added to 95% by weight of surfactant solution. The composition is mixed in a closed container until a homogeneous liquid is formed.

EXAMPLE 11

This example illustrates a cleaning composition of the present invention having a neutral pH. The composition components are:

Component	Weight %
Dodecyl/tridecylbenzene sulfonic acid	5.13
Sodium cumene sulfonate (40% aq. sol.)	6.80
Dipropyleneglycol-n-butyl ether	2.64
Sodium hydroxide	0.66
Ortho-phenylphenol	0.20
Sodium sulfate	0.99
Isopentane	5.00
Water, deionized	to 100
$\sum_{i=1}^{i=9} X_i P_i$	11.96 psia

The surfactant and hydrotrope are dissolved sequentially in the water. Ortho-phenylphenol is

dissolved into the ether cosurfactant and the resulting mixture is added to the surfactant solution with continued agitation. Sodium hydroxide is added slowly to obtain a pH of from about 5 to about 9 and sodium sulfate was added
5 under constant mixing at room temperature until homogeneous. The isopentane is cooled to about 5°C and is added to 95% by weight of surfactant solution. The composition is mixed in a closed container until a homogeneous liquid is formed.

10 EXAMPLE 12

The relationship between "the initial foam time" of compositions of the present invention and the type or mixture of water immiscible compound is demonstrated in this example. The compositions of Example 12 are prepared
15 using the ingredients listed in Table 1, in the manner described in Example 6.

"Time to initial foam" is determined using a blind sample evaluation and a single trigger sprayer (Model T85NDB, from Continental Sprayers) mounted on
20 either PET or glass containers. The trigger sprayer is rinsed before each use and each composition is sprayed 12 times before an evaluation. "Time to initial foam" is determined for each composition by spraying each composition a total of 6 times at a vertical target at a
25 distance of 6 inches and averaging the determinations. "Time to initial foam" is a measurement of the time from trigger actuation until the composition foams to obscure the target. The results are shown in TABLE 1.

TABLE 1

Material/Sample	12A	12B	12C	12D	12E	12F
Alkylbenzene sulfonic acid (BIOSOFT S100)	3.90%	3.90%	3.90%	3.90%	3.90%	3.90%
Dipropyleneglycol- n-butyl ether	2.00%	1.95%	1.90%	1.85%	1.80%	1.75%
Citric acid (anhydrous)	2.0%	2.0%	2.0%	2.0%	2.0%	2.0%
Magnesium Oxide	0.25%	0.25%	0.25%	0.25%	0.25%	0.25%
Water (deionized)	86.85%	86.90%	86.95%	87.00%	87.05%	87.10%
Isopentane	1.935%	2.432%	2.979%	3.582%	4.254%	5.0%
ISOPAR™C	3.065%	2.568%	2.021%	1.418%	0.748%	0.0
Avg. Time to initial foam (sec)	5.6	4.7	3.65	2.65	2.0	1.6
$\sum_{i=1}^n X_i P_i$	6.39 psia	7.50 psia	8.62 psia	9.73 psia	10.84 psia	11.96 psia

The results in TABLE 1 show that the "foam time" of the composition varies with the vapor pressure of the composition. Thus, the time in which a foam appears after application may be controlled by the varying amount and vapor pressure of the self-foaming agent.

EXAMPLE 13

A. Unthickened Self Foaming Laundry Pretreatment Composition, Examples 13A-13C.

These compositions are made from the following ingredients:

	Example 13A	Example 13B	Example 13C
Deionized Water	to 100	to 100	to 100
Sodium pareth-4- sulfate, 60% ^a	6.0	8.0	8.4
Tallowamine EO5 ^b	3.6	2.4	0
Soyamide DEA ^c	0	0	2.52
Propylene glycol	5.0	5.0	7.0
Dipropylene glycol n- propyl ether ^d	4.0	4.0	3.6
Sodium tetraborate decahydrate	1.0	2.0	2.8
Sodium sulfate	0	1.0	0
Enzyme ^e	0.3	0.3	0.3
Isopentane	4.25	4.25	4.25
Isopar C	0.75	0.75	0.75
$\sum_{i=1}^{i=N} X_i P_i$	10.84	10.84	10.84

^a= Sterol 460CS, from Stepan Chemical; ^b= Varonic T205, from Witco Chemical; ^c= Alkamide DIN 295S, from Rhone-Pouenc; ^d= Dowanol DPnP, from The Dow Chemical Company; ^e= Optimase APL-440, from Solvay Enzymes.

- The compositions are prepared by adding the water and the sodium pareth-4 sulfate to a 4 liter stainless steel beaker, then adding the propylene glycol and sodium tetraborate until they are dissolved. In Examples 13A And
- 5 13B, the dipropylene glycol n-propyl ether is then added. In the case of Example 13C, the sodium sulfate is added at this time and the dipropylene glycol n-propyl ether and Soyamide DEA are separately mixed and added. In all
- 10 cases, the enzyme is then added and mixed until uniform. The resulting mixture is transferred to polyethylene terphthalate bottles, and the isopentane and Isopar C are added. The bottles are then capped and the composition mixed by manual agitation.

B. Thickened Laundry Pretreatment Compositions 13D-13G

Example	13D	13E	13F	13G
Deionized Water	to 100	to 100	to 100	to 100
Sodium Pareth-4 sulfate, 60% ^a	8.4	6.1	0	0
Soyamide DEA ^b	2.52	3.93	0	0
C11.5 Alkylbenzene sulfonic acid ^c	0	0	3.5	3.5
Dinonylphenol EO7 ^d	0	0	3.6	3.6
Propylene glycol	7.0	6.67	5.0	5.0
Dipropylene glycol n-	3.6	4.33	4.0	4.0

propyl ether ^a				
Sodium tetraborate decahydrate	2.8	0.5	1.25	1.0
Sodium Hydroxide, 50% aqueous	0.22	0	0	0.11
Triethanolamine (85% in diethanolamine)	0	0	2.0	2.0
Enzyme ^c	0.3	0.3	0.3	0.3
Xanthan gum ^g	0	0.15	0.15	0
Acrylic acid polymer ^h	2.0	0	0	1.0
Isopentane	4.25	4.25	4.25	4.25
Isopar C	0.75	0.75	0.75	0.75
$\sum_{i=1}^{12} X_i P_i$	10.84	10.84	10.84	10.84

^aSteol 460CS, from Stepan Chemical; ^bAlkamide DIN 295S, from Rhone-Pouenc; ^cBiosoft S100, from Stepan Chemicals; ^dIgepal DM430, from Rhone-Poulenc; ^eDowanol DPnP, from The
5 Dow Chemical Company; ^fOptimase APL-440, from Solvay Enzymes; ^gKelzan T, from Kelco; ^hAcusol 820, from Rohm & Haas.

Examples 13D and 13E are prepared by adding the water and the sodium pareth-4 sulfate to a 4 liter stainless steel beaker, then adding the propylene glycol, sodium tetraborate and acrylic acid polymer (Ex. 13D) or xanthan gum (Ex 13E) until they are dissolved. In Example 13D, the sodium hydroxide is then added. The dipropylene glycol n-propyl ether and Soyamide DEA are separately mixed and added. The enzyme is then added and mixed until uniform. The resulting mixture is transferred to polyethylene terphthalate bottles, and the isopentane and Isopar C are added. The bottles are then capped and the composition mixed by manual agitation.

Examples 13F and 13G are prepared in the same manner, with the C11.5 alkylbenzene sulfonic acid and triethanol amine substituting for the sodium pareth-4 sulfate, and the dinonylphenol EO7 substituting for the soyamide DEA.

WHAT IS CLAIMED IS:

1. A composition in the form of a microemulsion comprising:

5 (a) water,

(b) from about a 0.5 to about 10 weight percent of a water immiscible compound or mixture thereof, the water immiscible compound(s) having a vapor pressure which satisfies the relationship

10
$$3 \text{ psi} < \sum_{i=1}^{i=y} X_i P_i < 14.7 \text{ psi.}$$

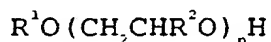
wherein y represents the number of water immiscible compounds in component (c), x_i represents the mole fraction of a water immiscible compound i in component (c), and P_i represents the vapor pressure of said water-immiscible compound i at 22° C;

15

(c) from about 0.2 to about 20 weight percent of a surfactant; and

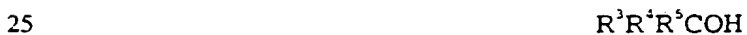
(d) from about 0.1 to about 20 weight percent of a cosurfactant selected from the group consisting of

20 1) an alkylene glycol ether of the formula



where R^1 is C_1 - C_6 alkyl, R^2 is H or methyl and n is a number from 1 to 3,

2) an alcohol of the formula



where R^3 , R^4 and R^5 are each either H or C_1 - C_7 alkyl provided that the total

number of carbon atoms in R^3 , R^4 and R^5 is 3 to 7, and

3) mono- and di-carboxylic acids having an equivalent weight of up to 88;

5 the relative amounts of components (a)-(d) being selected such that the composition is in the form of a liquid oil-in-water microemulsion.

2. The composition of Claim 1 wherein the water immiscible compound(s) have a vapor pressure which
10 satisfies the relationship

$$5\text{psi} < \sum_{i=1}^{i=n} X_i P_i < 12.5 \text{ psi.}$$

at 22°C.

3. The composition of Claim 1 wherein the major portion of the water immiscible compound is a C_5 - C_7 ,
15 aliphatic hydrocarbon.

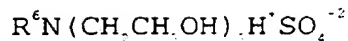
4. The composition of Claim 3 wherein the C_5 - C_7 aliphatic hydrocarbon is isopentane, n-pentane, or a mixture thereof.

5. The composition of Claim 3 wherein the
20 water immiscible compound further contains a C_8 - C_{12} aliphatic hydrocarbon.

6. The composition of Claim 6 wherein the surfactant is anionic or cationic.

7. The composition of Claim 6 wherein the
25 anionic surfactant is a metal salt of a C_{10} - C_{16} alkylbenzene sulfonate, a metal salt of a C_{12} - C_{30} alkyltoluene sulfonate, or a mixture thereof.

8. The composition of Claim 7 wherein the cationic surfactant is of the formula



where R^6 is C_{16-18} alkyl.

9. The composition of Claim 2 wherein the cosurfactant is dipropyleneglycol n-butyl ether,
5 dipropyleneglycol n-propyl ether, propyleneglycol n-butyl ether, propyleneglycol n-propyl ether, tripropyleneglycol methyl ether, or a mixture thereof.

10. The composition of Claim 9 wherein the water immiscible compound is present in the composition in
10 an amount of from about 1 to about 8 weight percent.

11. The composition of Claim 9 wherein the cosurfactant is present in the composition in an amount of from about 0.5 to about 10 weight percent.

12. The composition of Claim 11 wherein the
15 surfactant is present in an amount of from about 0.5 to about 10 weight percent.

13. The composition of Claim 1 further comprising an antimicrobial agent.

14. The composition of Claim 1 further
20 comprising a hydrotrope.

15. The composition of claim 1 wherein component (c) is a mixture of an anionic surfactant and a nonionic surfactant.

16. A packaged composition comprising:

25 (I) a container having a non-pressurized reservoir and an opening, said reservoir containing a composition in the form of a microemulsion having:

(a) water,

(b) from about a 0.5 to about 10 weight percent of a water immiscible compound(s) having a vapor pressure which satisfies the relationship

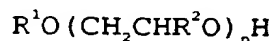
$$3 \text{ psi} < \sum_{i=1}^{i=y} X_i P_i < 14.7 \text{ psi.}$$

wherein y represents the number of water immiscible compounds in component (c), X_i represents the mole fraction of a water immiscible compound i in component (c), and P_i represents the vapor pressure of said water-immiscible compound i at 22°C;

(c) from about 0.2 to about 20 weight percent of a surfactant; and

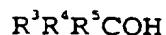
(d) from about 0.1 to about 20 weight percent of a cosurfactant selected from the group consisting of

1) an alkylene glycol ether of the formula



where R^1 is C_1 - C_6 alkyl, R^2 is H or methyl and n is a number from 1 to 3,

2) an alcohol of the formula



where R^3 , R^4 , and R^5 are each either H or C_1 - C_7 alkyl provided that the total number of carbon atoms in R^3 , R^4 , and R^5 is 3 to 7, and

3) mono- and di-carboxylic acids having an equivalent weight of up to 88;

the relative amounts of components (a)-(d) being selected such that the composition is in the form of a liquid oil-in water microemulsion, said opening being fitted with a

dispensing apparatus which is actuated from a closed to an open position. Wherein when said apparatus is activated it dispenses said composition in the form of a liquid, non-foamed spray.

INTERNATIONAL SEARCH REPORT

International Application No
PLI/US 97/00371

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C11D17/00 C11D3/18

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	GB 2 190 681 A (COLGATE PALMOLIVE CO) 25 November 1987 see claims; examples ---	1,6,7, 9-14
A	WO 94 23012 A (DOW CHEMICAL CO) 13 October 1994 see claims 1-7; examples ---	1,3-7,9
A	US 5 462 690 A (RHINESMITH ROBERT) 31 October 1995 see claims see claims ---	1,6,9
A	WO 94 02109 A (DOWBRANDS INC) 3 February 1994 see the whole document ---	1-16

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

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28 May 1997

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 97/00371

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>DATABASE WPI Section Ch, Week 9326 Derwent Publications Ltd., London, GB; Class C07, AN 93-208017 XP002031808 & HU 62 649 A (SZIASZ SZOLGALTATO ES IPARI ALTALANOS) , 28 May 1993 see abstract -----</p>	1,3

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 97/00371

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
GB 2190681 A	25-11-87	US 5075026 A	24-12-91
		AT 957 U	26-08-96
		AU 597367 B	31-05-90
		AU 7313887 A	26-11-87
		BE 1001742 A	27-02-90
		BR 8702610 A	23-02-88
		CA 1302194 A	02-06-92
		CH 676854 A	15-03-91
		DE 3716526 A	26-11-87
		DK 168958 B	18-07-94
		FR 2599046 A	27-11-87
		HK 60094 A	08-07-94
		LU 86888 A	22-11-88
		NL 8701215 A	16-12-87
		SE 503826 C	16-09-96
		SE 8702084 A	22-11-87
		SG 20994 A	10-06-94
		TR 25009 A	01-09-92
		US 5082584 A	21-01-92
		US 5076954 A	31-12-91
WO 9423012 A	13-10-94	AU 6528694 A	24-10-94
		BR 9405958 A	12-12-95
		EP 0692021 A	17-01-96
		JP 8508536 T	10-09-96
		US 5597792 A	28-01-97
US 5462690 A	31-10-95	US 5587357 A	24-12-96
WO 9402109 A	03-02-94	AU 4786893 A	14-02-94
		CN 1087380 A	01-06-94
		EP 0652739 A	17-05-95
		ES 2075820 A	01-10-95
		FI 950373 A	27-01-95
		HU 70703 A	30-10-95
		JP 7509513 T	19-10-95
		NO 950309 A	27-01-95
		PL 307296 A	15-05-95
		US 5602091 A	11-02-97